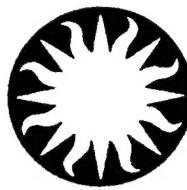




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ENERGY DISTRIBUTION OF FAST NITROGEN ATOMS IN THE NIGHTTIME TERRESTRIAL THERMOSPHERE

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15. ABSTRACT (Maximum 200 words)

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Energy distribution of fast nitrogen atoms in the nighttime terrestrial thermosphere

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Abstract

Energetic nitrogen atoms play a significant role in the chemistry and thermal balance of the thermosphere. The nocturnal energy distribution is calculated and the importance of including the center of mass motion is demonstrated. The center of mass motion gives rise to an extended component of fast atoms. The calculation of the energy distribution includes the loss of energetic nitrogen atoms in reactions with molecular oxygen. The density of energetic atoms and their energy distribution are strong functions of local time, altitude, and latitude.

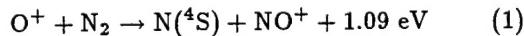
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1. Introduction

Reactions of energetic N(⁴S) atoms with O₂ molecules may be a significant source of nitric oxide molecules in the atmosphere [Solomon, 1983; Lie-Svendsen *et al.*, 1991]. The sources of energetic N(⁴S) atoms in the daytime have been explored by Gerard *et al.* [1991, 1993] and by Shematovich *et al.* [1991, 1992], who calculated the resulting energy distribution and pointed to their importance in producing NO in the daytime thermosphere. Sharma *et al.* [1993] argued that the reaction of N(⁴S) atoms with O₂ yields NO molecules in highly excited rotational and vibrational levels and proposed that they are responsible for the anomalous infrared emission features detected in the spectrum of the dayglow by Smith and Ahmadjian [1993] and Armstrong *et al.* [1994]. Nocturnal sources of energetic N(⁴S) atoms also exist. Scattered solar radiation at 58.4 nm and 30.4 nm from the extended helium geocorona produces O⁺ ions by photoionization of oxygen atoms and N₂⁺ ions by photoionization of nitrogen molecules and scattered solar Lyman alpha radiation at 121.6 nm photoionizes NO molecules to produce NO⁺ ions. Some O⁺ ions are produced by charge transfer with oxygen atoms of H⁺ ions transported from the dayside. The ions undergo chemical reactions leading to fast N(⁴S) atoms. The fast N(⁴S) atoms, reacting with O₂, may contribute to the nocturnal rotationally enhanced NO emission which overlaps the thermal emission spectrum produced by the excitation of NO by oxygen atom impact.

2. Sources of Nonthermal Nitrogen Atoms

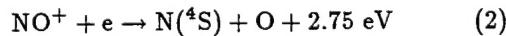
The O⁺ ions may react with N₂,



to produce N(⁴S) atoms with kinetic energies up to 0.74 eV with a rate coefficient k_1 given as a function of temperature T by [Roble *et al.*, 1987]

$$k_1 = \{1.533 - 0.592(T/300) + 0.086(T/300)^2\} \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}.$$

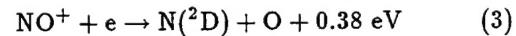
The NO⁺ ions present in the nocturnal atmosphere undergo dissociative recombination



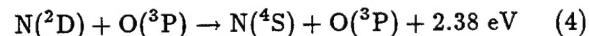
with a rate coefficient

$$k_2 = 9.2 \times 10^{-8}(T_e/300)^{-0.85} \text{ cm}^3 \text{ s}^{-1}$$

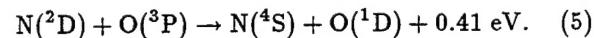
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with a rate coefficient $k_3 = 3.3 \times 10^{-7}(T_e/300)^{-0.85} \text{ cm}^3 \text{ s}^{-1}$ [Roble *et al.*, 1987] where T_e is the electron temperature. Reactions (1), (2), and (3) are the major sources of nitrogen atoms at night. Dissociative recombination of N₂⁺ is negligible [Tohmatsu, 1990] in comparison. Following reaction (3), energetic N(⁴S) atoms are produced by the reactions

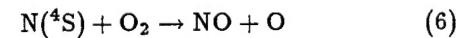


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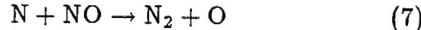
The sum of the rate coefficients k_4 and k_5 of reactions (4) and (5) has been measured at thermal energies [Piper, 1989; Fell *et al.*, 1990]. No experimental data are available on the branching ratio of the individual reactions but Bates [1989] has tentatively concluded from an examination of the potential energy curves of NO calculated by Michels [1979] that at room temperature, process (5) is strongly favored.

To calculate the source function of energetic N(⁴S) atoms we adopted the ion and electron densities in the international reference ionosphere [Rawer *et al.*, 1978] and the neutral particle densities and temperatures in the MSIS-86 [Hedin, 1987] model atmospheres for quiescent nighttime conditions near solar maximum. The altitude profiles of the ion and electron densities are reproduced in Figure 1. The N₂⁺ densities are too small to appear in the figure. We make the arbitrary assumption that in reaction (1) none of the energy is taken up in internal modes of NO⁺. The fast atoms lose energy mostly in elastic collision with the ambient atmosphere. For these exploratory investigations we adopted a hard sphere potential to describe the elastic collisions [Andersen and Shuler, 1964; Oppenheim *et al.*, 1977]. The fast atoms are lost in reactions with O₂



to form NO. The kinetics of reaction (6) have been explored by Gilibert *et al.* [1993, 1995] and Duff *et al.* [1994] who calculated the classical trajectories on the NO₂ potential energy surfaces and determined the fraction of collisions that are reactive. In our calculations we adopted the cross sections of Duff *et al.* [1994] which show reaction occurring at kinetic energies of the nitrogen atoms above 0.43 eV. Below 0.43 eV, the nitrogen atoms are preferentially removed by

the reaction with NO,



which has a rate coefficient of $2 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$, independent of temperature [Koshi *et al.*, 1990]. The thermalization by elastic collisions is rapid compared to the time scales of the transport phenomena that are important in determining the density of thermal N(⁴S) atoms, which we took from the MSIS model atmospheres. Superimposed on the thermal background are the nascent N(⁴S) atoms which react with O₂ as they slow down, producing NO. We took the NO densities from the empirical model of Smith *et al.* [1993].

3. Energy Distribution

Shematovich *et al.* [1991, 1992] took account of the numerous daytime sources of energetic nitrogen atoms and solved the Boltzmann equation to obtain the daytime steady-state energy distribution. They assumed that the energy spectra of the sources could be represented as delta functions centered at the specific initial energies, determined by the exoergicities of the processes that produce the nitrogen atoms. These representations of the source spectra are valid in the center of mass frame. In the laboratory frame they are considerably broadened because the angle between the velocity of the center of mass and the relative velocity of the separating systems varies randomly. Figure 2 is the Newton diagram for the dissociative recombination reactions (2) and (3). If \mathbf{W}_N and \mathbf{W}_O are the velocities of the nitrogen and oxygen atoms, respectively, in the center of mass frame and \mathbf{V}_{cm} is the velocity of the center of mass, the velocities of O and N in the laboratory frame are $\mathbf{V}_O = \mathbf{W}_O + \mathbf{V}_{cm}$ and $\mathbf{V}_N = \mathbf{W}_N + \mathbf{V}_{cm}$. Their magnitudes are $\{W_O^2 + V_{cm}^2 + 2W_O V_{cm} \cos\Theta\}^{1/2}$ and $\{W_N^2 + V_{cm}^2 - 2W_N V_{cm} \cos\Theta\}^{1/2}$ where Θ is the angle between \mathbf{W}_O and \mathbf{V}_{cm} . The velocities of O and N are related by the conservation of linear momentum so that $M_O \mathbf{W}_O + M_N \mathbf{W}_N = 0$ where M_O is the mass of the oxygen atom and M_N is the mass of the nitrogen atom. The circles in Figure 2 denote the loci of the atom velocities in the center of mass frame. The spread in velocities obtained by adding vectorially the velocity of nitrogen in the center of mass frame to the velocity of the center of mass produces a width τ in the source spectrum of the order $(\Delta k_B T)^{1/2}$ where Δ is the exoergicity. The detailed analysis is presented in the appendix. For the nitrogen atoms resulting from reactions (1), (2), (4), and (5), the widths are large compared to $k_B T$.

At low altitudes, reaction (1) is unimportant as a source of N. Figure 3a shows the result of an accurate calculation of the source functions at an altitude of 130 km for reactions (2), (3), and (4) for a temperature of 550 K, compared to Gaussian functions with a half-width of $k_B T$, assuming that reaction (5) is negligible. Figure 3b is identical except that reaction (4) is replaced by reaction (5). The inclusion of the center of mass motion has a dramatic effect on the source spectrum. We calculated the energy distribution function $P(E)$ by solving the Boltzmann equation

$$\frac{\partial}{\partial t} P(E) = 0 = \int_0^\infty B(E', E) P(E') dE' - \kappa(E) P(E) + Q(E, t) - \alpha(E, t) P(E), \quad (8)$$

where the kernel $B(E', E)$ is the rate of transfer of kinetic energy from E' to E , $Q(E, t)$ is the source function of atoms with energy E , $\alpha(E, t)$ is the sink function that denotes the frequency of reactive collisions in which nitrogen atoms are lost, and

$$\kappa(E) = \int_0^\infty B(E, E') dE' \quad (9)$$

is the frequency of elastic collisions that remove atoms with energy E . The distribution function $P(E)$ is normalized so that $\int_0^\infty P(E) dE$ equals the density of nascent nitrogen atoms. The source and sink functions depend on the local time t , which is a parameter in (8). The kernel is a sum,

$$B(E', E) = \sum_j B_j(E', E), \quad (10)$$

of energy transfer rates $B_j(E', E)$ occurring through collisions of the energetic nitrogen atoms with the constituents N₂, O₂ and O of the ambient atmosphere. For hard sphere potentials the kernel may be expressed in analytical form [Anderson and Shuler, 1964; Oppenheim *et al.*, 1977]. The sink term describes the loss of nitrogen atoms in reactions (6) and (7). The effects of including the center of mass motion in the source function are illustrated in Figure 4 which presents the steady state distribution in the atmosphere at an altitude of 130 km corresponding to the source functions in Figure 3. The peaks are smoothed out by the inclusion of the center of mass motion and an increase by orders of magnitude occurs in the high energy tail of the distribution function above 2 eV. In Figure 5 we present the source functions arising from reactions (1) through (5) at altitudes of 130 km,

160 km and 200 km at local midnight. For Figure 5a we assume that only reaction (4) quenches $N(^2D)$ and for Figure 5b we assume that only reaction (5) quenches $N(^2D)$. Below 130 km the energetic nitrogen atoms are produced mostly by dissociative recombination and by quenching $N(^2D)$ but at higher altitudes the reaction of O^+ with N_2 is the major source. Figure 6 gives the energy distributions $P(E)$ of the nascent atoms at 19.20 local time at altitudes of 130 km, 160 km, and 200 km, and at a latitude of -28.0° , and a longitude of 102.4° . The thermal Maxwellian distributions are also shown. Because the nitrogen atoms live longer at higher altitudes where molecular oxygen is a relatively less abundant constituent, and because the bath gas has a higher temperature, the thermal component of the distribution persists to larger energies. The choice of the quenching channel (4) or (5) is of little consequence. Figure 7 is similar to Figure 6 but for local midnight. Even at midnight there is present a significant abundance of energetic atoms. Figure 8 shows the energy distribution at 19.30 local time for a different latitude, -48.0° , and longitude, 105° , based on the internal reference ionosphere data. The different initial ion composition is reflected in the energy distribution function. Figures 6-8 show that energetic atoms are present through the night and that their density and energy distribution are strong functions of the geophysical parameters.

The reactions of the energetic atoms populate excited rotational and vibrational levels of NO . The resulting infrared emission [Sharma *et al.*, 1993] has been seen during the daytime [Smith and Ahmadjian, 1993; Armstrong *et al.*, 1994], and Armstrong *et al.* [1994] have suggested it may be the source of a weak rotationally excited component present in quiescent nighttime spectra. Quantitative calculations will be needed to determine whether or not the supply of energetic atoms is sufficient to explain the observed intensities.

Appendix

At time t the energy spectrum of the source function $Q(E, t)$ of the fast nitrogen atoms in the laboratory frame (LF) can be written as the product

$$Q(E, t) = Q_0(t)q(E), \quad Q_0(t) = \int_0^\infty Q(E, t)dE, \quad (11)$$

where $Q_0(t)$ is the total rate of production of fast atoms and $q(E)$ is the normalized source function of the atoms in the LF, such that $\int q(E)dE = 1$.

Transforming the distribution function from the center of mass frame (CMF) to the LF, we obtain

$$q(E) = \int \delta [E - M_N(\mathbf{V}_{cm} + \mathbf{W}_N)^2/2] \times F(\mathbf{V}_{cm})d\mathbf{V}_{cm}f_0(\mathbf{W}_N)d\mathbf{W}_N, \quad (12)$$

where $f_0(\mathbf{W}_N)$ is the velocity distribution function of the fast nitrogen atoms in the CMF and $F(\mathbf{V}_{cm})$ is the distribution function of the center of mass velocities. If $f_0(\mathbf{W}_N)$ is isotropic (see Figure 2) and the reaction generates a monoenergetic flux of atoms with velocity W_* ,

$$q = \int \delta \left[E - M_N \left(\frac{V_{cm}^2 + W_*^2}{2} + V_{cm}W_*\cos\Theta \right) \right] \times F(V_{cm})V_{cm}^2dV_{cm}2\pi\sin\Theta d\Theta. \quad (13)$$

The velocity of the fast atomic particles in the CM frame W_* depends on the exoergicity of the chemical reaction Δ :

$$W_* = \left[\frac{2M_O\Delta}{M_N M} \right]^{1/2}, \quad (14)$$

where $M = M_N + M_O$. Integration with respect to the angle variable Θ leads to a restriction on the magnitude of CM velocities allowed for the given E and W_* :

$$V_{min} \leq V_{cm} \leq V_{max}, \quad (15)$$

where

$$V_{min} = \left| W_* - (2E/M_N)^{1/2} \right|, \quad (16)$$

$$V_{max} = W_* + (2E/M_N)^{1/2} \quad (17)$$

The velocity distribution of the CM motion is Maxwellian. Using it we obtain an analytical expression for the source function:

$$q(E) = \left(\frac{M}{2\pi kT} \right)^{1/2} \frac{1}{M_N W_*} \times \left\{ \exp \left[-\frac{MV_{min}^2}{2kT} \right] - \exp \left[-\frac{MV_{max}^2}{2kT} \right] \right\}. \quad (18)$$

The source energy distribution function $q(E)$ may now be calculated using the dependence of W_* on the exoergicity Δ . The result is

$$q(E) = \frac{(\frac{M_O}{M_N})^{1/2} + (\frac{M_N}{M_O})^{1/2}}{(\pi\Delta kT)^{1/2}} \exp \left[-\frac{ME + M_O\Delta}{M_N kT} \right] \times \sinh \left[\frac{2(E\Delta M_O M)^{1/2}}{kTM_N} \right]. \quad (19)$$

For large values of the ratio Δ/kT the position of the maximum of $q(E)$ and the energy width τ of the source function in the LF are given approximately by

$$E_{\max} \sim \frac{M_O}{M_N + M_O} \Delta, \quad (20)$$

and

$$\tau \sim \frac{2(M_O M_N)^{1/2}}{M_N + M_O} (\Delta kT)^{1/2}. \quad (21)$$

A similar result can be obtained from the analyses of *Whipple* [1974] and *Wallis* [1978].

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Figure 1. The altitude profiles of the ion and electron densities.

Figure 2. Newton diagram relating the velocity vectors of the N and O atoms and the center of mass frame with those in the laboratory frame. V_{cm} is the velocity of the center of mass.

Figure 3. The thermal spectrum at 550 K is compared to the calculated distribution, taking into account the center of mass motion. The peaks on the right are due to reaction (2) and on the left to reaction (4) in Figure 2a and reaction (5) in Figure 2b. The center of mass motion causes a broadening of the distribution to a few tenths of an electron volt.

Figure 4. Energy distribution function $P(E)$ of nascent N atoms from the source function of Figure 2a at midnight and 130 km altitude as a function of the translational energy from a thermal source (broken line) and taking account of the center of mass motion (solid line). The Maxwell distribution at a local temperature of 560 K is shown as the dashed-dotted line.

Figure 5. reference Figure 5. Energy spectrum of the source function $Q(E, t)$ at 130 km, 160 km and 200 km at midnight. Figures. 5a and 5b assume reactions (4) and (5), respectively, for the deactivation of $N(^2D)$ atoms by $O(^3P)$ atoms.

Figure 6. Energy distribution function $P(E)$ of nascent $N(^4S)$ atoms at 130 km, 160 km and 200 km. The solid lines assume reaction (4) and the broken lines assume reaction (5) for the deactivation of $N(^2D)$ atoms by $O(^3P)$ atoms. The geophysical parameters are 19.20 local time, latitude -28° , and longitude 102.6° . The dashed-dotted curves indicate the Maxwellian velocity distributions.

Figure 7. As in Figure 6 except the local time is midnight.

Figure 8. As in Figure 6 except the local time is 19.30, the latitude is -48° and the longitude 105° .

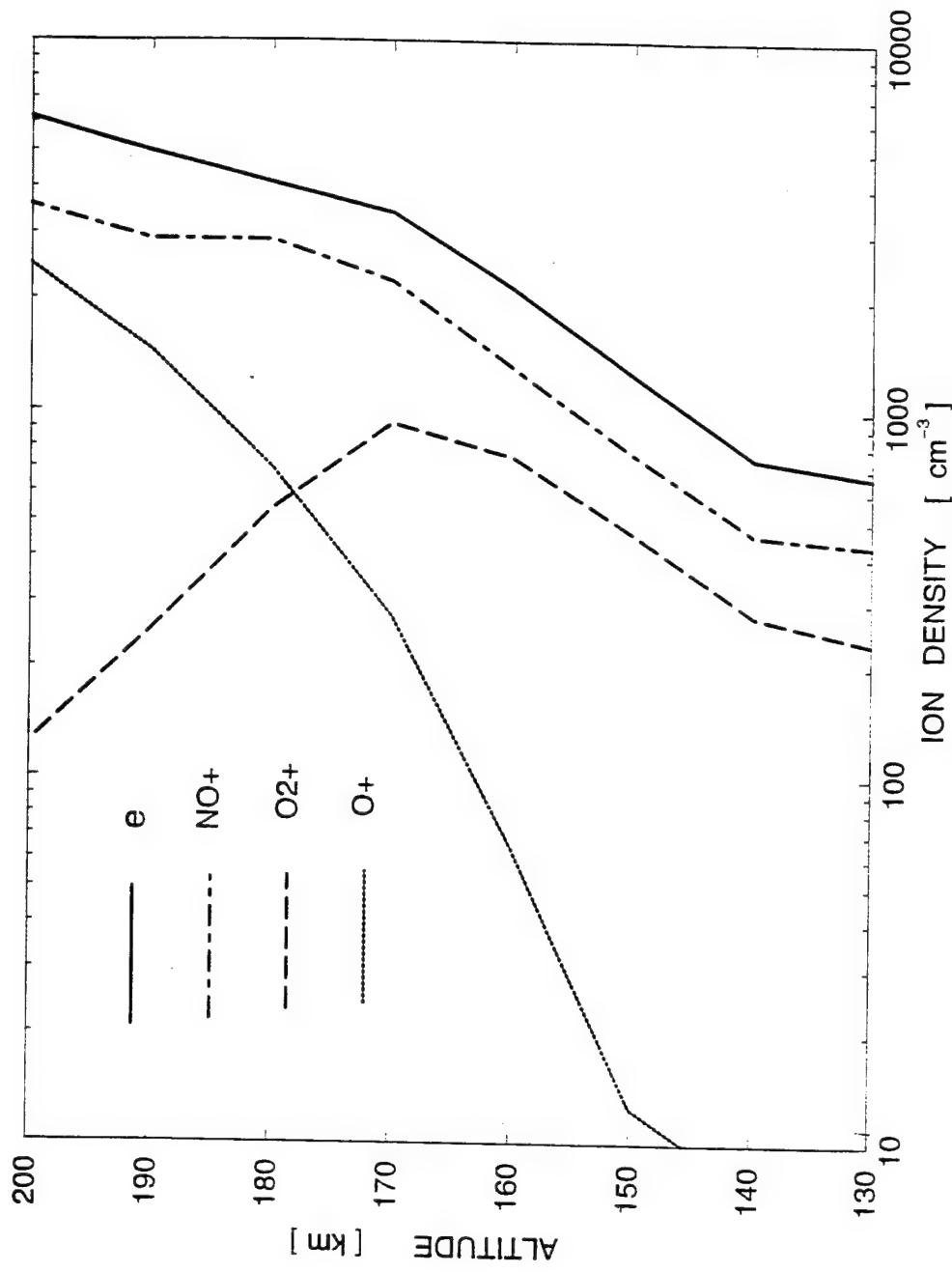


Fig 1

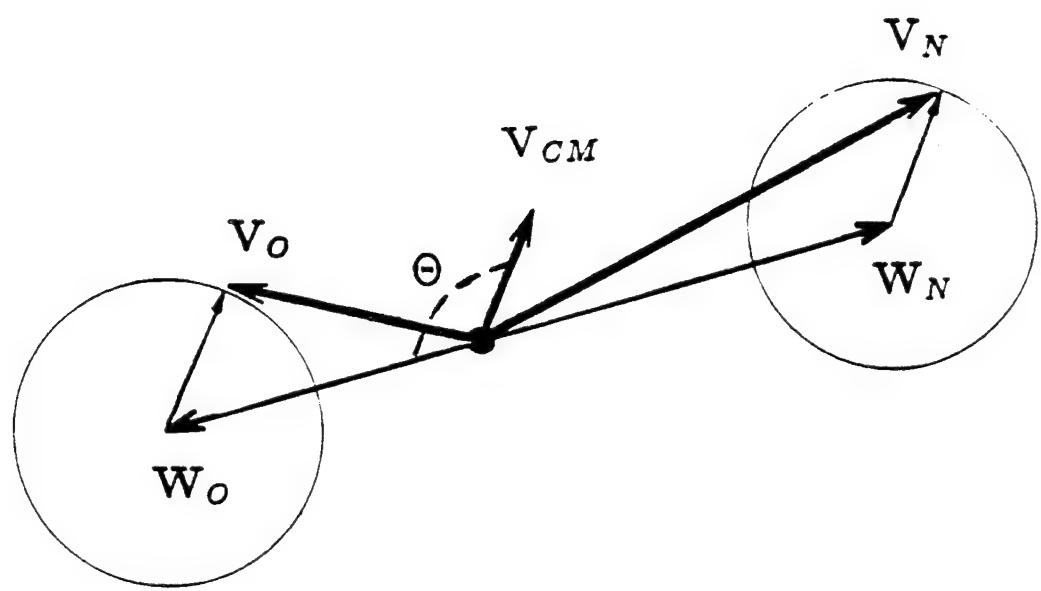


Fig2

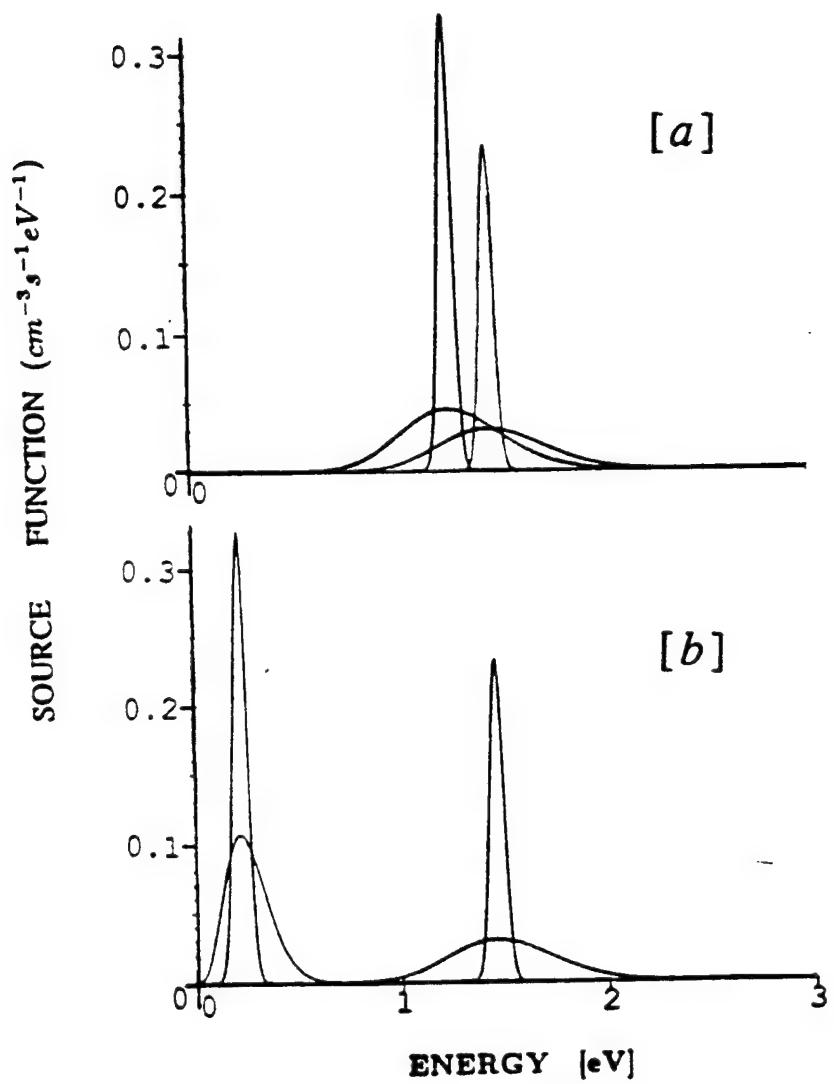


Fig3

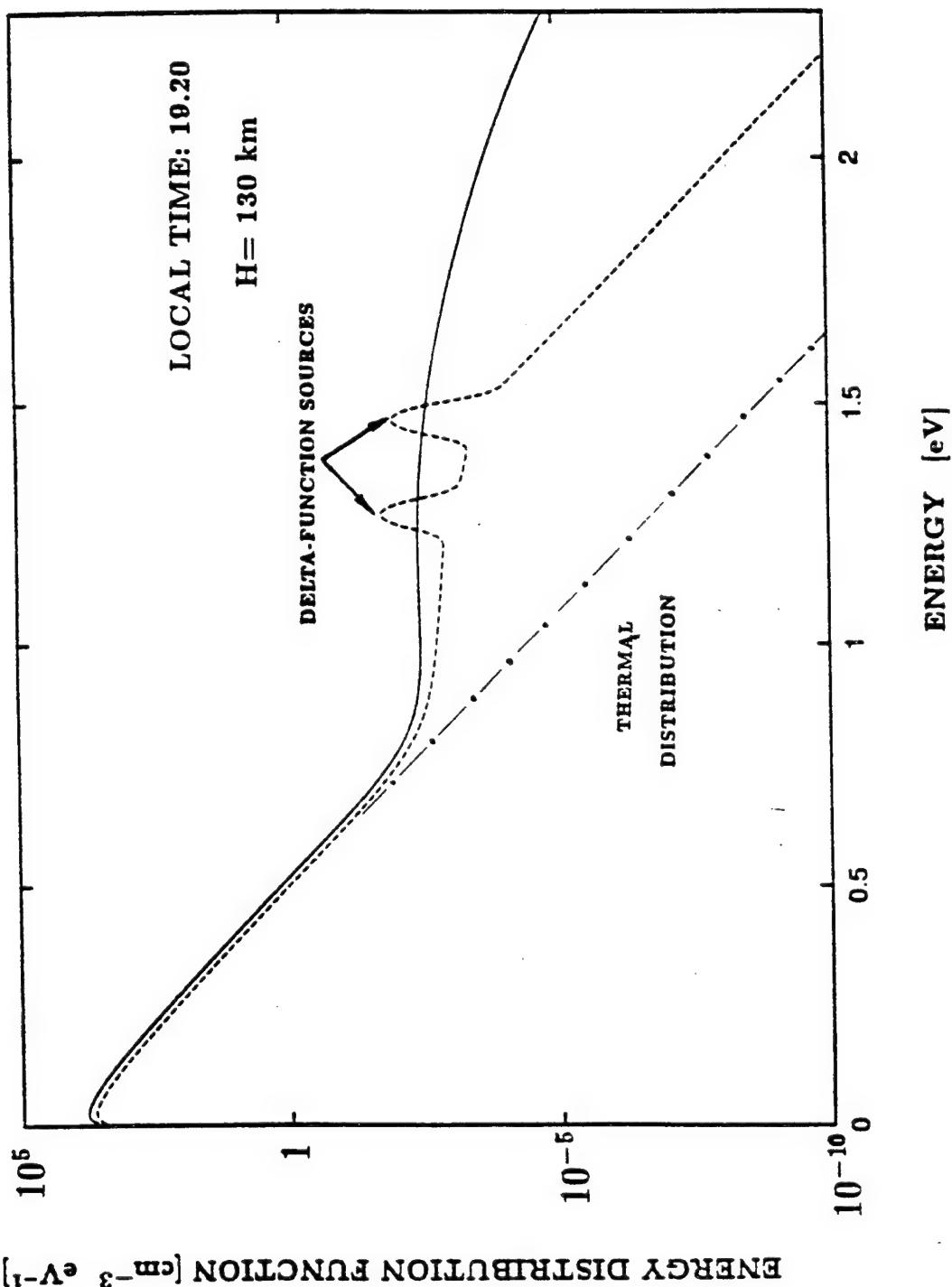


Fig 4

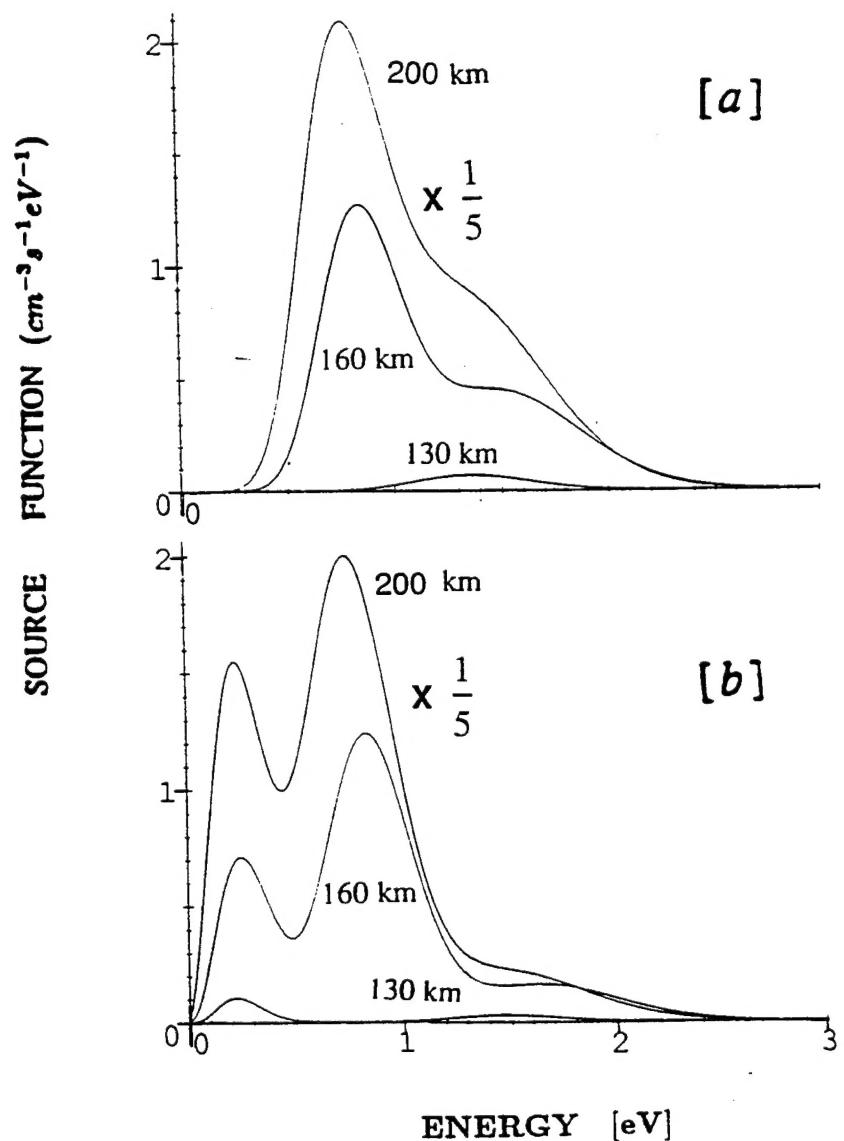


Fig5

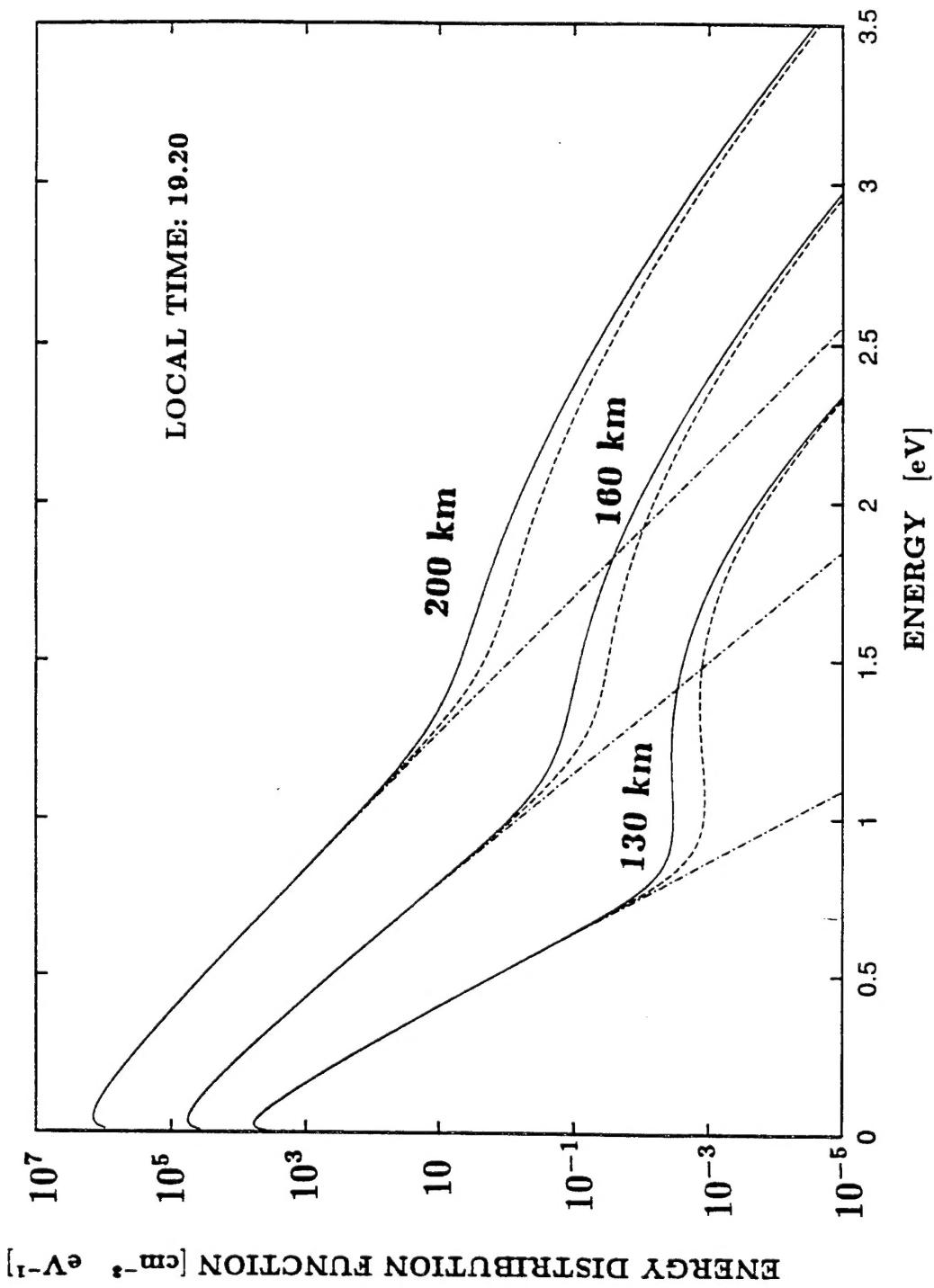


Fig6

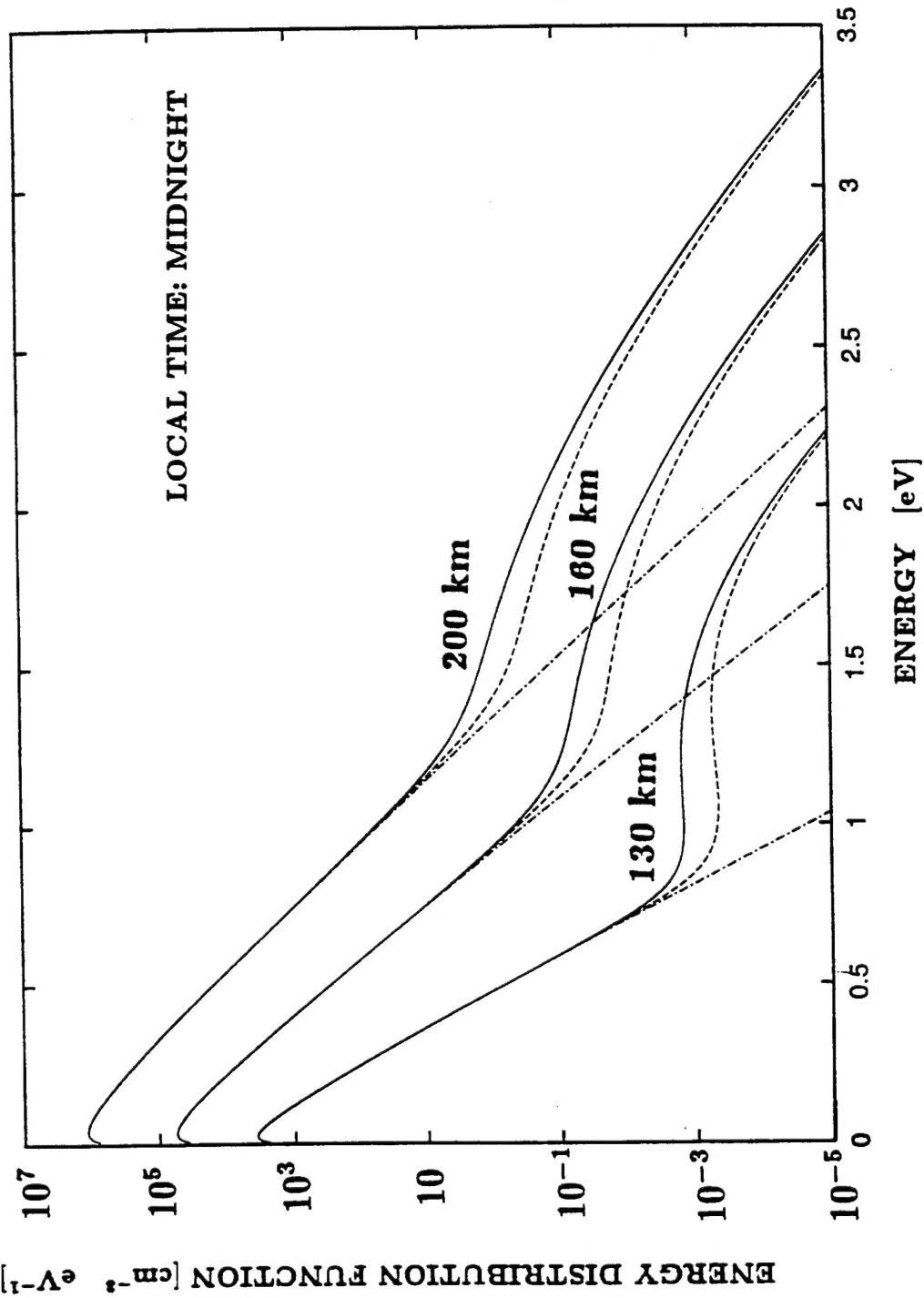


Fig 7

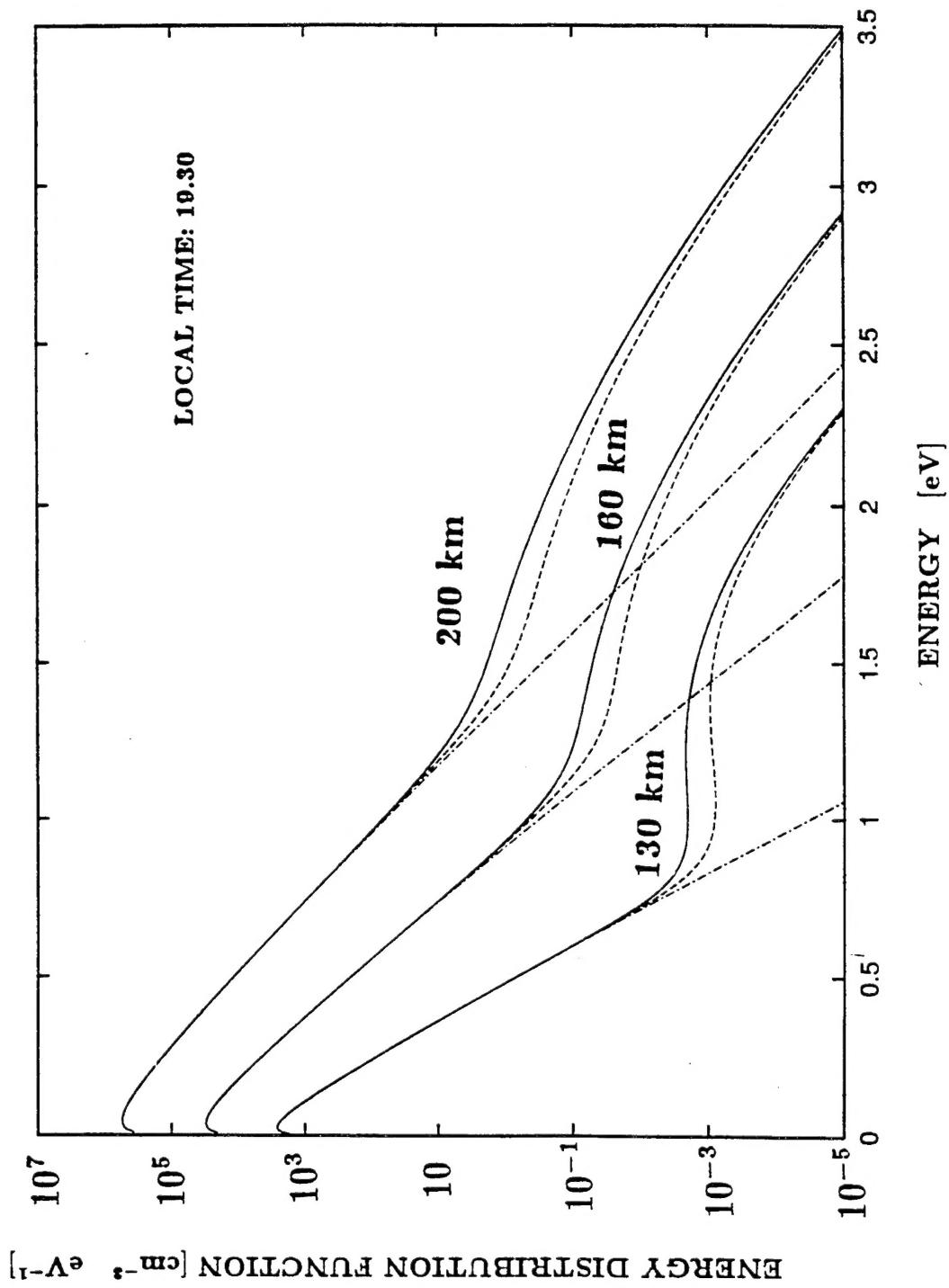


Fig 8